Advanced Lab Course: Optically detected magnetic resonance of photo-excited triplet states in nitrogen vacancy centers of diamond

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1. Introduction

1.1. Before we start

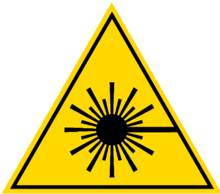
Optically detected magnetic resonance (ODMR) in diamond nitrogen-vacancy (NV) centers is a noninvasive high-resolution-high-sensitivity magnetometry technique applicable to a wide temperature range [1]. Its experimental implementation is tied to a variety of challenges, in particular if the object under investigation requires cryogenic cooling. The development of a low temperature scanning magnetometry setup is an ongoing project at the Institute of Physics. The setup used in this *advanced lab course* serves as a development and test platform for the future cryogenic version, and is subject to continuous adaptations. The following instructions serve merely as guiding principles and have to be taken with a grain of salt. The final setup and measurement plan will be discussed within the scope of the colloquium.

1.2. Safety MATTERS

The most important aspect of your laboratory work is your safety. Within the scope of this experiment, you will encounter the following hazards:

Danger! *Every work environment bears its hazards and dangers. This applies in particular to laboratory settings where working procedures and equipment are flexible and cannot be standardized. A strong sense for potential risks and a high focus at work is in order.*

Tripping danger! *The devices used in this experiment are very sensitive to light and the laboratory must be kept dark. Mind proper housekeeping to avoid tripping hazards.*



Laser radiation! *The 515 nm diode-pumped solid-state laser (DPSS) used in this experiment emits up to 80 mW of light. The laser class is 3B, i.e. eye exposure to the direct or reflected beam is harmful [2]. Keep your head well above the optical table! Remove reflecting accessories like watches, jewelry, etc. from your arm wrists! Reduce the laser output as much as possible! Wear laser safety goggles if possible!*

Electrical danger! *The 3D vector magnet is powered by a 80 V/5 A power supply. Do not touch powered electrodes! Do not unplug or switch off the device while the magnet is at field! Do not reboot the controlling PC while the magnet is at field!*

Magnetic field! *The Helmholtz coils develop a magnetic field up to 5 mT. Do not insert magnetic materials into the coil space!*

2. Preparing for the Experiment

The following section gives a guideline to a proper preparation for this experiment along with a minimal but non-exhaustive literature recommendation:

2.1. The NV-Center in diamond

The nitrogen vacancy (NV) center in diamond is well documented in references [3, 4, 5]. For a proper preparation, inform yourself about the following aspects:

- **Formation & structure:** How are NV centers in diamond formed? What is their structure? What is their symmetry?
- **Electronic structure:** Which are the possible electron configurations in the NV center, and which one do we employ in ODMR? What is the electronic structure of the relevant NV center and does this relate to its symmetry? Identify the electronic, phononic and magnetic substructure and their energetic hierarchy.
- **Optical transitions:** Which are the possible electronic transitions and which ones do we study in ODMR. What is the role of dipole selection rules? How do the electronic transition interact with the magnetic and phononic structure?
- **Magnetic subsystem:** What is the minimal Hamiltonian of the spin-triplet subsystem subject to a magnetic field? Which terms of the Hamiltonian respect the symmetry of the ideal NV center, and which terms don't? What is the energy level diagram with and without magnetic field? Under which circumstances can we neglect magnetic field components perpendicular to the axis of the NV defect?
- **ODMR:** How does the application of a microwave field affect the population of the magnetic subsystem? How does this indirectly affect the electronic subsystem and its transitions?
- **Crystal orientation:** Based on the above, develop a strategy to measure the orientation of an NV-center. How can this help us to determine the exact orientation of the diamond crystal?
- **Zeeman splitting:** Based on the above, develop a strategy to measure the Zeeman splitting in the NV-center. Along which axis should the magnetic field be preferentially applied?

- **Rabi oscillations:** Sufficiently coupled two level systems will undergo *Rabi oscillations* [6]. What is the source for the level coupling in our case and how does the frequency depend on a measurable quantity of that source? How does a system react to a so called π-*pulse*?

2.2. Methods and Devices

While the experimental setup described in section 3.1 is under constant development, the measurement principle will not change. Familiarize yourself with the most important components and the procedures employed in this experiments. Discuss the details of the actual setup with your tutor.

- **Optics:** Familiarize yourself with each optical element of the experimental setup. What is its working principle and how is it operated? What is it needed for in this particular experiment?

- **Compact Spectrometer:** Familiarize yourself with the working principle of a modern compact spectrometer. How does it have to be operated to obtain a meaningful spectrum? Which spectral range is relevant in this experiment?

- **APD:** Familiarize yourself with the working principle of an avalanche photo diode (APD). What is the meaning of *dark count rate* and *saturation*? What is *gated photon counting* and how can it be used to improve measurement stability against systematic errors?

- **AOM:** Familiarize yourself with the working principle of an acousto optic modulator (AOM). What does it do and how does it work? For which parts of the experiment do we need it?

- **Trigger:** Familiarize yourself with the concept of triggered data acquisition. What is a *TTL* signal? What is its role for *gated photon counting*?

- **ODMR:** Familiarize yourself with the working principle of optically detected magnetic resonance. What is is its advantage with respect to conventional detection schemes? How do we actually measure ODMR in our experiment?

- **LabView:** Equipment and data acquisition will be controlled by virtual interfaces (VIs) written in *LabView*. Knowledge of LabView programming is not necessary, but it will be helpful to familiarize yourself with some basic features.

3. The Experimental Setup

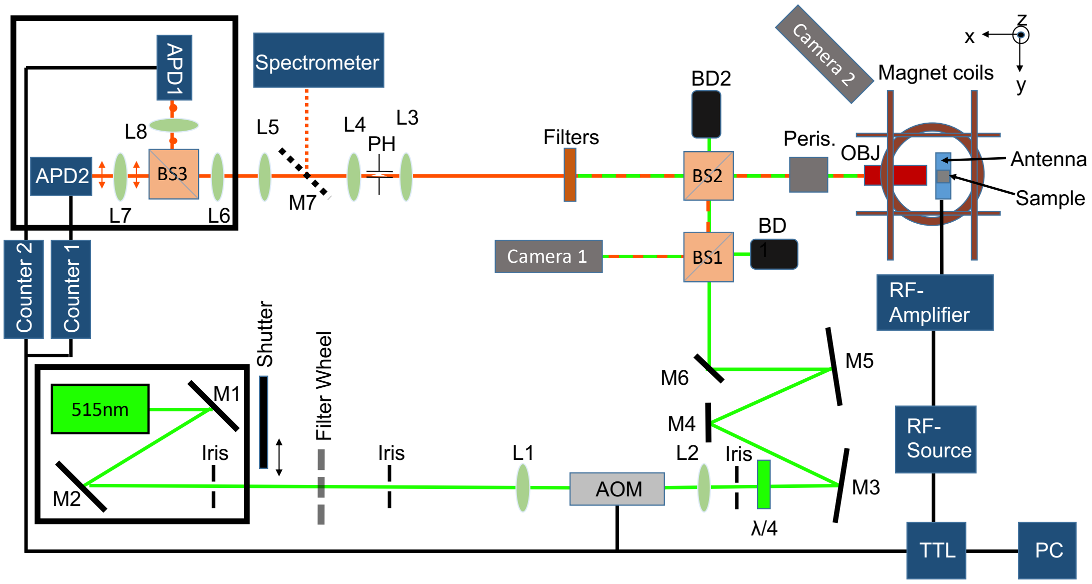


Figure 3.1.: The optical table. M: silver coated mirror; L: lens; BD: beam dump; BS beam splitter; OBJ: microscope objective; PH: pin hole (15 μm diameter); Peris: periscope.

3.1. Hardware

The system used in this experiment (see Fig. chapter 3) is under constant development. This section gives a rough introduction to its principal components. The actual experimental implementation may deviate from this description. For further information, please consult the operation manuals and discuss the final setup with your tutor.

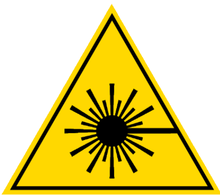
3.1.1. Optical table

The optical table consists of a nonmagnetic stainless steel plate, with a raster of M6 thread holes for mounting optical elements, that is fixed onto a massive granite plate. This structure is floating on air pressurized vibrational dampeners. Weak changes in the force distribution on the optical table will tilt the table and redistribute the air pressure to re-level the surface.

Do not drop heavy weights onto the table. Do not lean against the table.

Laser system

The laser used in this setup is a class 3B *Omicron Phoxx 515-80* diode pumped solid state (DPSS) laser, emitting continuous light at a wavelength of 515 nm with a maximum Power of 80 mW. It is fully controlled by Labview and has no physical control elements. Two mirrors M1 and M2 direct the laser beam onto the optical laser path (green) that is defined by two irises. For alignment, the power can be reduced by up to 4 orders of magnitude by a filter wheel in front of the laser's housing box. For complete blockage of the beam, a metal shutter is provided.



Eye exposure to the direct or reflected beam is harmful! Reduce the laser output as much as possible! Wear laser safety goggles!

Acousto optic modulator (for Rabi oscillations)

In order to chop the laser intensity for the Rabi experiment, an acousto optic modulator (AOM) in combination with two lenses L1 and L2 is provided. The key benefit of the AOM as compared to a direct modulation of the laser is the higher extinction/contrast ratio (diode modulation typically $1:200$, AOM typically $< 1:1000$). In continuous laser mode, the AOM is operated on the zero order beam. In pulsed mode, the first order diffraction is used. All other orders are dumped into an iris aperture. The selection of the desired diffraction order is performed by two free standing 2 inch diameter mirrors M3 and M5. The on/off state of the AOM is controlled by the TTL signal generated by a fast pulse generator PC card (*SpinCore PulseBlaserESR-PRO500*). The delay between the TTL signal and the AOM response is ~ 650 ns and needs to be accounted for in the measurement settings.

The microscope

Mirror M6 couples the laser into the microscope path (red/green dashed). To increase the mechanical stability of this system and to ease the alignment process, most parts are integrated into a cage system. All other components offer a stable alignment as well and should not be touched. Beamsplitter BS1 and BS2 along with a periscope and a microscope objective OBJ focus the laser onto the diamond sample. The microscope objective type *Mitutoyo Plan Apo NIR HR 100x* with a 10 mm working distance, a 0.70 numeric aperture and a 2 mm focal length, collects the back-reflection of the laser as well as the photoluminescence signal from the diamond. BS2 and BS1 guide parts of this signal into the microscope path and onto CMOS camera 1 *DCC 1545M*, which allows the user to view the sample. An additional CMOS camera 2 (*Thorlabs DCC 1545M/C*) provides a side view of the sample and is used to monitor the laser spot on the sample. Beam dumps BD1 and BD2 absorb unwanted deflections.

Waveguide diamond stack

The diamond sample is cut along the (1 0 0) plane, and glued into a radio frequency (RF) antenna. This stack is mounted onto the micro positioning system with the [1 0 0] diamond axis along the beam axis and the thin layer of NV centers facing away from the microscope objective.

A Labview controlled *Stanford Research Systems (SRS) SG384* RF-generator produces microwaves up to 4 GHz at a power level of 16.5 dBm [7]. The output of this generator is fed into an RF switch whose output depends on the TTL signal level. The signal is further amplified by a 35 dB *microwave amps AM7-2.6-3-33-33* amplifier, followed by an RF waveguide that couples the microwaves into the antenna. The waveguide is terminated by a 50 Ohm load.

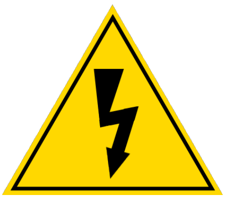
Micro positioning system

The waveguide diamond stack is mounted onto the three axis micro positioning system *Attocube ANC 300* that operates via three *slip-stick* piezo drives. These allow to position the diamond with sub-micrometer resolution over several millimeters of travel range. This allows for precise positioning of the diamond stack with respect to the microscope objective.

Magnetic coils

The waveguide diamond stack along with the micro positioning system is placed at the center of three Helmholtz coil pairs, providing a magnetic field strength of about 5 mT along each Cartesian coordinate x, y and z. The Helmholtz coils are powered by three fast corrector current- and voltage-controlled digital power supplies type FAST-PS 0580-400 that provide up to ±80 V at ±5 A (400 W max power).

Heating of the powered coils may lead to sample drift!



Do not touch powered electrodes! Do not unplug or switch off devices in operation! Do not reboot the controlling PC or terminate the LabView program while the magnet is at field!

Do not insert magnetic materials into the coil space!

Wavelength filters

Most of the laser back-reflection as well as the photoluminescence signal collected by the microscope objective OBJ is guided into the detection path (red). To remove the intense primary laser signal from the detection path, a filter is inserted. The following filters are available:

- *Thorlabs FELH550*, 550 nm long pass
- (690 ± 40) nm band pass

Compact spectrometer

A removable mirror M7 deflects the signal into a fiber coupler, with a fiber guiding the signal to a compact USB spectrometer *OceanOptics QE65Pro*. The Peltier cooler of the spectrometer's CCD chip is powered by an external supply and requires about 30 seconds to reach operating temperature ($\sim -17^\circ$ C). The spectrometer is operated via the *OceanView* software installed on the desktop of the control PC.

The removable mirror M7 is locked by a strong magnet! Insert the mirror slowly and with great care!

Never insert the mirror without a filter in the detection path! The spectrometer can be damaged by intense laser radiation!

Single photon counting module

Upon removal of mirror M7, the signal passes directly into a closed box containing beam splitter BS3 as well as two extremely sensitive avalanche photo diodes (APD). BS3 acts as a polarizer and passes the horizontal light components towards APD2 (extinction ratio 1:1000), while the vertical light components are deflected to APD1 (extinction ratio 1:20). The APDs are integrated into a stabilized controlling electronics module (*Excelitas SPCM-AQRH-14*). Due to the dead time after a detection event, the APDs saturate at $\sim 15 \times 10^6$ million cps. Count rate linearity with respect to the actual photon rate can be expected until 10^6 cps. Each APD is read out by a *Stanford Research Systems (SRS) SR400* photon counter. Both counters are equipped with two input channels A and B, individually triggered by the TTL signal and read out by Labview.

Do not open APD housing! Block the entrance of the APD housing when not in use! Block the entrance of the APD housing when lights are on!

3.2. PC and Software

The hardware necessary for data acquisition is controlled by dedicated software installed on the measurement PC. Like the hardware, the software is under constant development and may alter over time. Their detailed function and operation will be discussed upon startup of the experiment. The main tasks of the PC are:

- To control the USB spectrometer and to acquire spectra (Software OceanView).
- To control and display the monitoring cameras (Software ThorCam).
- To start up individual device controls; to facilitate data acquisition and life data monitoring (Labview: simple control center).
- To control the positioning system (Labview: position control system).
- To set the laser power (Labview: phoxx main).

- To set the RF frequency and power (Labview: SG-384).
- To set the magnetic field (Labview: magnet power supply).
- To provide the TTL signal to the RF generator, the photon counter and the AOM (Labview: pulseblaster).
- To set the trigger command of the photon counter and read its value (Labview: photon counter).

3.3. Gated photon counting

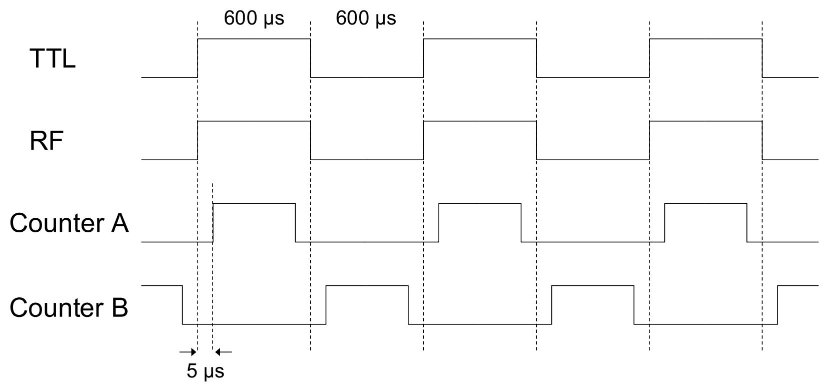


Figure 3.2.: The principle of gated photon counting.

ODMR measures the relative change of the NV-centers' photoluminescence (PL) emission upon sweeping an applied RF signal. In order to measure the relative change in PL intensity with and without this RF field, *gated photon counting* is employed. The principle is shown in Fig. section 3.3. The fast pulse generator card of the PC (type *SpinCore PulseBlaserESR-PRO500*) generates a TTL signal of 1.2 ms period. This signal triggers the RF-switch that consequently changes its output state from *on* to *off* at the same frequency. The trigger also activates the counting channels A and B of the two individual photon counters. Channel A counts the detected photons upon the application of RF, while channel B counts the detected photons with the RF-generator off. A delay time of 5 μs accounts for possible settling times of the RF system. The relative intensity is formed as the quotient of count-rates A/B. The frequency of the TTL signal is much higher than typical variations of the background illumination, which compensates systematic errors on the timescale of the entire measurement.

4. Measurement plan

The laser should be used at a power setting of 50 % for all experiments.

The filter wheel can be used to further reduce the light intensity. You should always start with a fast measurement setting and estimate optimal parameters on that basis.

4.1. Optical alignment

Your advisor will introduce you to the optical components and assist you in the alignment process. Typically, the following alignment procedures will be necessary:

- Adjust diamond position and focus via piezo stage. Feedback signal is the image of camera 1 as well as the count rate on the APDs.
- Adjust coupling of the signal into the spectrometer's fiber via lens L4. Use PL spectrum as feedback signal.
- Adjust coupling of the signal into the APDs via lens L4. Use count rate on APDs as feedback signal.
- In rare cases, adjust the spatial filter components. Use count rate on APDs as feedback signal.

4.2. PL spectrum of NV centers

With mirror M7 inserted and the low pass filter in, optimize the signal on the spectrometer and take a photoluminescence spectrum of the NV centers. The signal to noise ratio should be on the order of 1000:1. For meaningful data interpretation, refer to reference [5]. Questions to be answered:

- What are the energies of the zero phonon lines and the phonon satellites of NV^0 and NV^- centers.
- What are their uncertainties and how are these affected by systematic errors?

A strategy for analysis:

- Introduce the spectrum.
- Formulate a model.

- Select a fit function based on this model.
- Perform the fit and discuss its significance.
- Discuss the validity of the model and what needs to be improved.

4.3. ODMR basics

With mirror M7 removed, optimize the signal on the APDs. Using the APDs, take an ODMR spectrum at zero magnetic field and determine the transversal zero field splitting of the NV centers in this diamond crystal. Apply a magnetic field of > 0.5 mT along each axis individually and record the respective ODMR spectrum. How do these differ for different directions of the magnetic field? Which is the preferential axis for Zeeman splitting measurements and why? Find a compromise between data statistics and data acquisition time such that you can fulfill your tasks within the given time (good data vs. a lot of data).

4.4. Zeeman splitting

Step the magnetic field along the axis of choice from -3 mT to 3 mT while recording the ODMR spectrum at every step. Mind that close to zero magnetic field, the magnetic step size should be finer to resolve the zero field splitting.

Questions to be answered:

- What is the appropriate fit function to fit a single ODMR spectrum? Which error should be used?
- What is the appropriate fit function to fit the entire Zeeman series? Which errors should be used?
- Determine the zero field splitting parameters D and E as well as the gyromagnetic ratio g.

4.5. Crystal orientation

Apart from some misalignment, the diamond [1 0 0] axis is pointing towards the microscope objective and therefore more or less along the magnetic x-axis. In this experiment, ODMR shall be used to determine the exact orientation of the crystal with respect to the magnetic coordinate system. First, set the magnetic field amplitude to 3 mT while rotating the field vector around the x axis in steps of about 10 degrees. Record an ODMR spectrum at every step. Second, based on the findings of the rough angular scan, select angular regions for which you perform angular scans with higher precision. Repeat step one and two for the y and/or z axis.

Questions to be answered:

- What is the appropriate fit function to fit the entire angular series? Which errors should be used?
- What is azimuthal orientation of the diamond with respect to the magnetic yz-plane? What are the appropriate errors?
- What is the out of plane tilt of the diamond? What are the appropriate errors?
- How are the results compatible with the Zeeman amplitudes of the individual ODMR branches?
- How are the results compatible with the intensity of the individual ODMR branches?
- Based on your finding, find all magnetic field directions along which the magnetic field is parallel to an NV axis.

4.6. Rabi oscillations (if time and setup allows)

Consult with your mentor if and how the experiment shall be carried out.

1. Based on the finding of the last section, align the magnetic field (~ 3 mT) parallel to an NV axis.
2. Using the settings of Tab. table 4.1, measure the ODMR signal while varying the RF pulse time between 11 ns and 1.9 µs in 40 ns steps.
3. Repeat the measurement for 3 different RF power settings (min. -11 dBm at SG384 output).

Questions to be answered:

- What are the Rabi frequencies as a function of microwave power?
- What are the spin coherence times T_2 as a function of microwave power?

Settings Photon Counter	Value
Mode	Single APD gated
Gate A&B width	0.5 us
Gate A delay	0.65 us
Gate B delay	2.65 us
Cycles	4000000

Settings PulseBlaster	Value
Sequence	Rabi Oscillation
Init time	11 ns
RF-Time	is set automatically
Read-out	500 ns
Wait for Ref.	2 us
Wait	2 us

Table 4.1.: Parameters used to observe Rabi oscillations.

A. Bibliography

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